

## Claims

[c1] CLAIMS:

1. A curable composition, comprising:

- (a) an epoxy resin and curing agent therefor, wherein said epoxy resin is essentially free of bromine atoms;
- (b) a flame retardant additive essentially free of phenolic groups and of epoxy groups, wherein said flame retardant is a condensation product of (i) a brominated phenol or a mixture of brominated phenols with (ii) a cyanuric halide;
- (c) a thermoplastic resin; and
- (d) a cyanate ester.

[c2] 2. The curable composition of claim 1, wherein said flame retardant additive has a bromine content greater than 20%.

[c3] 3. The curable composition of claim 1, wherein said flame retardant additive is 1,3,5-tris(2,4,6-tribromophenoxy)triazine.

[c4] 4. The curable composition of claim 1, wherein said flame retardant additive is 2,2'-[(1-methylethylidene)bis[(2,6-dibromo-4,1-phenylene)oxy]]bis[4,6-bis[(2,4,6-tribromophenyl)oxy]-1,3,5-triazine].

[c5] 5. The curable composition of claim 1, wherein said flame retardant additive is soluble in toluene at a concentration of greater than 15 g/100ml of toluene at a temperature of 50 ° C.

[c6] 6. The curable composition of claim 1, wherein said epoxy resin is a glycidyl ether resin or a mixture of glycidyl ether resins containing, on average, greater than 2 epoxy groups per molecule.

[c7] 7. The curable composition of claim 1, wherein said epoxy resin is a mixture of:  
(a1) an epoxy resin containing on average less than or equal to 2 glycidyl groups per molecule; and  
(a2) an epoxy resin containing greater than 2 glycidyl groups per molecule.

[c8] 8. The curable composition of claim 1, wherein said thermoplastic resin has a

Tg greater than 120 ° C.

- [c9] 9. The curable composition of claim 1, wherein said thermoplastic resin has a dissipation factor of less than 0.010 measured at 1 MHz at room temperature.
- [c10] 10. The curable composition of claim 1, wherein said thermoplastic resin has been directly isolated from solution after polymerization.
- [c11] 11. The curable composition of claim 1, wherein said thermoplastic resin is a poly(phenylene ether).
- [c12] 12. The curable composition of claim 11, wherein said poly(phenylene ether) has a weight average molecular weight ranging from about 3,000 to 35,000 g/mol.
- [c13] 13. The curable composition of claim 11, wherein said poly(phenylene ether) has a weight average molecular weight ranging from about 3,000 to 35,000 g/mol.
- [c14] 14. The curable composition of claim 11, wherein said poly(phenylene ether) has been melt processed at a temperature ranging from about 200 ° to 350 ° C.
- [c15] 15. The curable composition of claim 11, wherein said poly(phenylene ether) is hydroxy functional.
- [c16] 16. The curable composition of claim 1, wherein said thermoplastic resin is one or more of a poly(phenylene ether) or a poly(styrene- co -maleic anhydride).
- [c17] 17. The curable composition of claim 1, wherein said thermoplastic resin is a reaction product of a poly(phenylene ether) and a peroxide.
- [c18] 18. The curable composition of claim 1, wherein said thermoplastic resin is a reaction product of a poly(phenylene ether), a peroxide, and a bisphenol.
- [c19] 19. The curable composition of claim 1, wherein said thermoplastic resin is a polyimide.
- [c20] 20. The curable composition of claim 1, wherein the curable composition further comprises one or more of an organic reinforcement, an inorganic

reinforcement, or a filler.

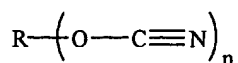
[c21] 21. The curable composition of claim 1, wherein the curable composition is essentially free of homopolymers of styrene.

[c22] 22. The curable composition of claim 1, wherein the epoxy resin is a multifunctional glycidyl ether.

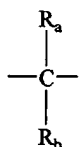
[c23] 23. The curable composition of claim 22, wherein said multifunctional glycidyl ether is selected from the group consisting of epoxidized phenol-formaldehyde novolacs, epoxidized cresol-formaldehyde novolacs, epoxidized alkylphenol-formaldehyde novolacs, epoxidized 1,1,1-tris(4-hydroxyphenyl)ethane, epoxidized 1,1,2,2-tetra(4-hydroxyphenyl) ethane, epoxidized phenol-dicyclopentadiene novolacs, and epoxidized phenol-benzaldehyde novolacs.

[c24] 24. The curable composition of claim 1, wherein the cyanate ester is selected from the group consisting of 1,3-dicyanatobenzene, 1,4-dicyanatobenzene, 2-tert-butyl-1,4-dicyanatobenzene, 2,4-dimethyl-1,3-dicyanatobenzene, 2,5-di-tert-butyl-1,4-dicyanatobenzene, tetramethyl-1,4-dicyanatobenzene, 4-chloro-1,3-dicyanatobenzene, 1,3,5-tricyanatobenzene, 2,2'-dicyanobiphenyl, 4,4'-dicyanobiphenyl, 3,3',5,5'-tetramethyl-4,4'-dicyanobiphenyl, 1,3-dicyanatonaphthalene, 1,4-dicyanatonaphthalene, 1,5-dicyanatonaphthalene, 1,6-dicyanatonaphthalene, 1,8-dicyanatonaphthalene, 2,6-dicyanatonaphthalene, 2,7-dicyanatonaphthalene, 1,3,6-tricyanatonaphthalene, bis(4-cyanatophenyl)methane, bis(3-chloro-4-cyanatophenyl)methane, bis(3,5-dimethyl-4-cyanatophenyl)methane, 1,1-bis(4-cyanatophenyl)ethane, 2,2-bis(4-cyanatophenyl)propane, 2,2-bis(3,3-dibromo-4-cyanatophenyl)propane, 2,2-bis(4-cyanatophenyl)-1,1,1,3,3,3-hexafluoropropane, bis(4-cyanatophenyl)ester, bis(4-cyanatophenoxy)benzene, bis(4-cyanatophenyl)ketone, bis(4-cyanatophenyl)thioether, bis(4-cyanatophenyl)sulfone, tris(4-cyanatophenyl)phosphate, and tris(4-cyanatophenyl)phosphate.

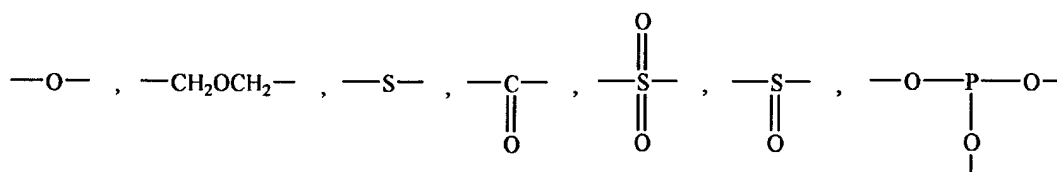
[c25] 25. The curable composition of claim 1, wherein the cyanate ester has the formula



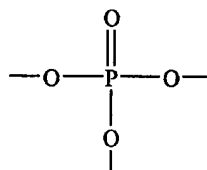
wherein R is an aromatic nucleus-containing residue which is selected from the group consisting of a residue derived from an aromatic hydrocarbon selected from the group consisting of benzene, biphenyl and naphthalene, a residue derived from a compound in which at least two benzene rings are bonded to each other by a bridging member selected from the group consisting of



wherein R<sub>a</sub> and R<sub>b</sub> are the same or different and each represents a hydrogen atom or an alkyl group containing 1 to 4 carbon atoms,



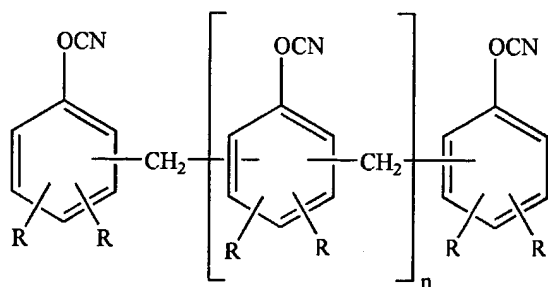
and



and a residue resulting from the removal of a phenolic hydroxyl group from a novolac-type or resol-type phenolic resin skeleton; said aromatic nucleus is optionally substituted by a substituent selected from the group consisting of alkyl groups containing 1 to 4 carbon atoms, alkoxy groups containing 1 to 4 carbon atoms, chlorine and bromine; n is an integer of 2 to 5; and the cyanate group is always directly bonded to the aromatic nucleus.

[c26] 26. The curable composition of claim 1, wherein the cyanate ester is a prepolymer of the cyanates esters of Claim 25, having a number average molecular weight of 400 to 6,000, and are formed by trimerizing the cyanate group of the cyanate esters.

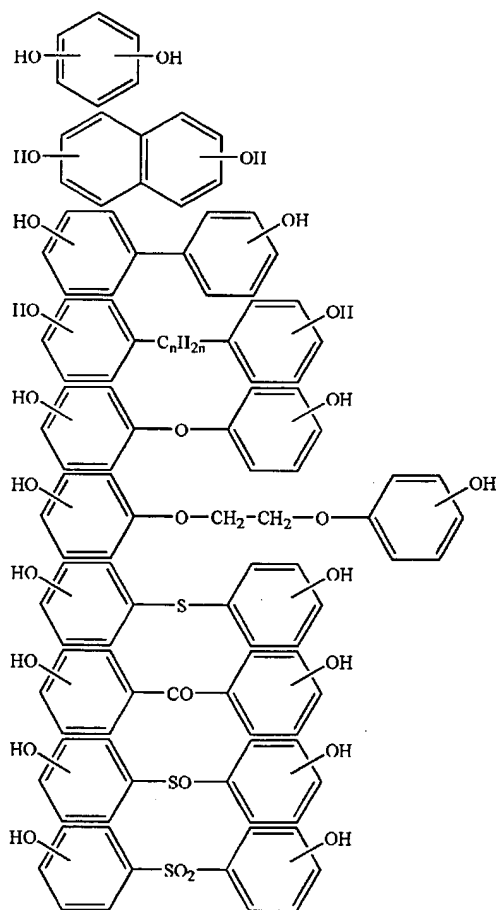
[c27] 27. The curable composition of claim 1, wherein the cyanate ester is a cyanate-group-containing phenol resin comprising a mixture of polymers represented by the formula



wherein  $n$  is 0 or an integer of 1 or more; and R's may be the same or different, and each R is a hydrogen atom or a methyl group, and containing 50% by weight or more in total of polymers having formula in which  $n$  is an integer of 1 to 3, the number average molecular weight of said phenol resin being 350 to 700 g/mole.

[c28]

28. The curable composition of claim 1, wherein the cyanate ester is a cyanic acid ester of an aromatic polycarbonate obtained by reacting an aromatic polycarbonate having one or two terminal hydroxyl groups with a cyanogen halide, wherein the aromatic polycarbonate is prepared from an aromatic dihydroxy compound represented by one of the following formulas:



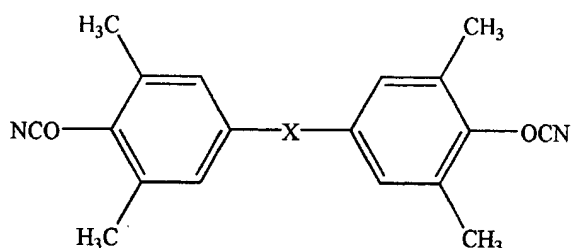
where  $n$  is an integer of 1-4, inclusive, or a mono-, di-, tri- or tetra- halogeno-

[c29]

$$\text{NCO-R-OCN}$$
[illegible]

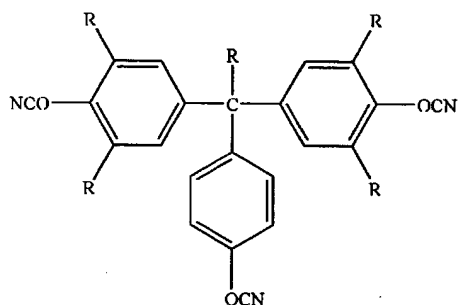
[c30]

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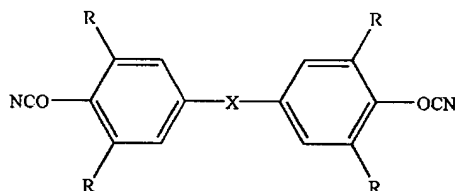


wherein X is methylene, isopropylidene, oxygen or divalent sulfur.

- [c31] 31. The curable composition of claim 1, wherein the cyanate ester is a blend of a tricyanate ester and a dicyanate ester, wherein the tricyanate ester has the structural formula:

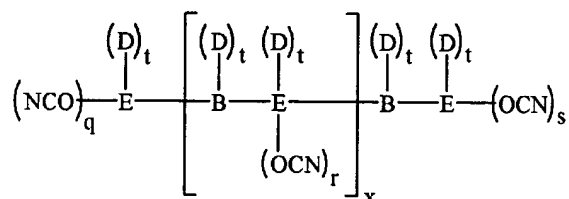


and the dicyanate ester has the structural formula:



wherein each R is H or methyl and is the same or different and wherein X is methylene, alkylidene having 2 to 4 carbon atoms, divalent oxygen, or divalent sulfur.

- [c32] 32. The curable composition of claim 1, wherein the cyanate ester is a polyaromatic cyanate having the formula



wherein: E is an aromatic radical;

B is a C<sub>7-20</sub> polycyclic aliphatic radical;

D is independently in each occurrence any nonactive hydrogen-containing substituent;

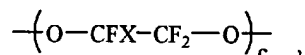
q, r and s are independently in each occurrence the integers 0, 1, 2, or 3; with

the proviso that the sum of q, r and s is greater than or equal to 2;  
 t is independently in each occurrence an integer of between about 0 and 4  
 inclusive; and  
 x is a number between about 0 and 5 inclusive.

- [c33] 33. The curable composition of claim 1, wherein the cyanate ester is a  
 fluorocarbon monocyanate having the structure  

$$\text{F}_3\text{C}(\text{CFX})_a\text{A}(\text{CFX})_b\text{CH}_2\text{OCN}$$

where A is



X is fluorine or perfluoroalkyl having 1 to 10 carbon atoms, a is 1 to 10, h is 1,  
 and c is 1 to 100.

- [c34] 34. The curable composition of claim 1, wherein the cyanate ester is a  
 fluorocarbon dicyanate having the structure  

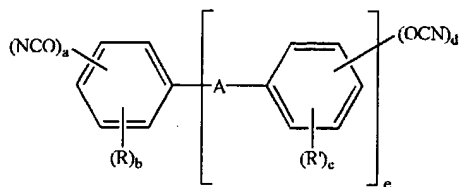
$$\text{NCOCH}_2(\text{CFX})_a\text{B}(\text{CFX})_b\text{CH}_2\text{OCN}$$

where B is (I) a carbon-to-carbon bond, in which case a is an integer of 1 to 30  
 and b is zero, or (II) B is  $[(\text{CFX})_d\text{O}(\text{CFX})_e]_f$ , in which case a and b are zero, d  
 and e are integers of 1 to 30, and f is an integer of 1 to 20, or (III) B is  
 $(\text{OCF}_2\text{-CFX})_g\text{O}(\text{CFX})_h\text{O}(\text{CFX-CF}_2\text{O})_i$

in which case a and b are 1, h is an integer of 1 to 10, and g and i are integers  
 of 1 to 100, or (IV) B is  
 $[(\text{CF}_2\text{CH}_2)_j(\text{CF}_2\text{-CFX})_k]_m$ ,

in which case a and b are integers of 1 to 10, j and k are integers whose ratio  
 j/k is 1/1 to 10/1, m is an integer of 1 to 100, and  $(\text{CF}_2\text{CH}_2)$  and  $(\text{CF}_2\text{-CFX})$   
 are randomly distributed units; and where X in all instances where it appears is  
 fluorine or perfluoroalkyl of 1 to 10 carbon atoms.

- [c35] 35. The curable composition of claim 1, wherein the cyanate ester has the  
 formula

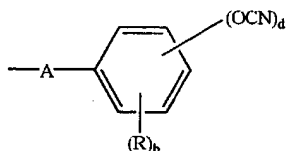


in which R represents hydrogen, halogen, linear or branched  $\text{C}_1\text{-C}_9$ -alkyl or  
 phenyl, two adjacent radicals R on the same nucleus together forming a



carbocyclic 5-membered or 6-membered ring or together and in conjunction with a hetero atom (O, S, N) forming a 5-membered or 6-membered heterocyclic ring, alkoxy radicals with 1 to 4 carbon atoms, alkoxy carbonyl radicals with 1 to 4 carbon atoms in the alkyl group;

R' has the same meaning as R or represents the group



where A is direct bond, a  $C_1-C_9$ -alkylene group optionally substituted by  $C_1-C_4$ -alkyl or phenyl, a cycloaliphatic or aromatic 5-membered or 6-membered ring, or a cycloaliphatic or aromatic 5-membered or 6-membered ring; a is a number from 0 to 5 where  $e = 1$  and a number from 2 to 5 where  $e = 0$ ;  $b = 5 - a$  where  $e = 1$  and  $6 - (a + d)$  where  $e = 0$ ;  $c = 5 - d$ ; d is a number from 0 to 5; e is the number 0, 1, 2 or 3; with the proviso that the sum of a and d ( $a + d$ ) always gives a number from 2 to 5.

[c36]

36.A curable composition, comprising:

- (a) an epoxy resin and curing agent therefor, wherein said epoxy resin is a glycidyl ether resin or mixture of glycidyl ether resins containing, on average, greater than 2 epoxy groups per molecule;
- (b) 1,3,5-tris(2,4,6-tribromophenoxy)triazine and/or 2,2'-[(1-methylethylidene)bis[(2,6-dibromo-4,1-phenylene)oxy]]bis[4,6-bis[(2,4,6-tribromophenyl)oxy]-1,3,5-triazine];
- (c) a poly(phenylene ether) resin; and
- (d) a cyanate ester.

[c37]

37.A curable composition, comprising:

- (a) an epoxidized cresol-formaldehyde novolac resin;
- (b) 1,3,5-tris(2,4,6-tribromophenoxy)triazine;
- (c) a poly(phenylene ether) resin having a number average molecular weight ranging from about 1,000 to 15,000 g/mol; and
- (d) a cyanate ester.

[c38]

38.A cured composition comprising a cured residue of a curable composition comprising:

- (a) an epoxy resin and curing agent therefor, wherein said epoxy resin is essentially free of bromine atoms;
- (b) a flame retardant additive essentially free of phenolic groups and of epoxy groups, wherein said flame retardant is a condensation product of (i) a brominated phenol or a mixture of brominated phenols with (ii) a cyanuric halide;
- (c) a thermoplastic resin; and
- (d) a cyanate ester.

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